Radioactivity - Radionuclides - Radiation
8th Nuclear Science Training Course with Nuclides.net
(Institute Jožef Stefan, Ljubljana, 13th-15th Sept. 2006)

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Nuclear Forensic science

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Content

1. Introduction

2. Analytical techniques

3. Examples of real cases

4. R & D

5. Conclusions

From illicit trafficking to nuclear forensics

1. Detection (border control, airport, police/intelligence)
2. Categorisation (nuclear material or radioactive source)
3. Nuclear forensic investigations

Aim of the identification of unknown nuclear material

- **Intended use** → Isotopic composition
- **Origin** → Isotope/elemental ratios, impurities, ...
- **Last legal owner** → Plant records, database
- **Smuggling route** → Non-radioactive traces

Nuclear forensic samples analysed at ITU

– 1992-1997

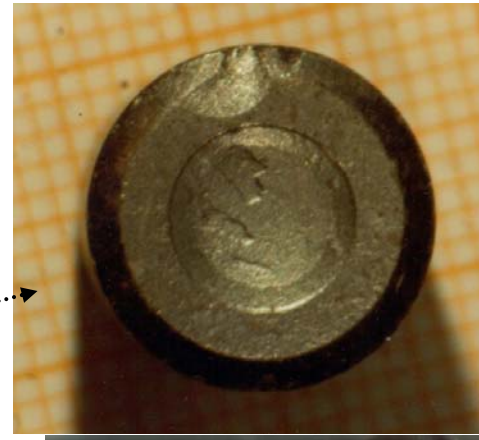
- 21 seizures:

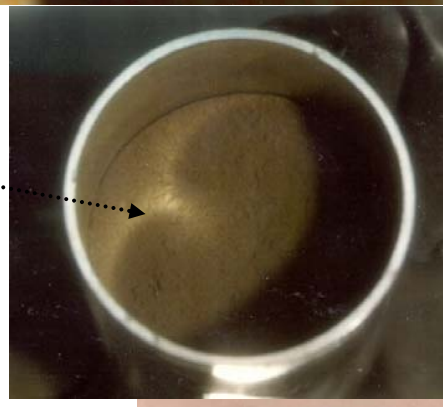
  nat.U, LEU fuel pellets, HEU, Pu,

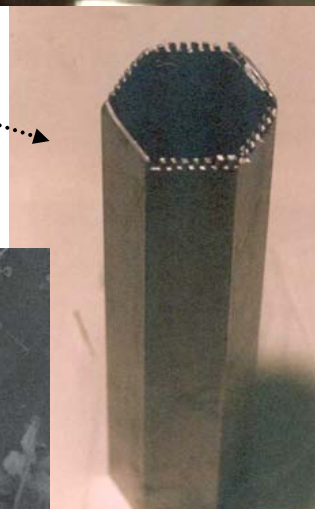
  contaminated scrap metal, ...

– 1997-today

- accidental release, theft, ...
- new seizures in 2004 and 2006







"Diagnostic principle"

On-site categorisation

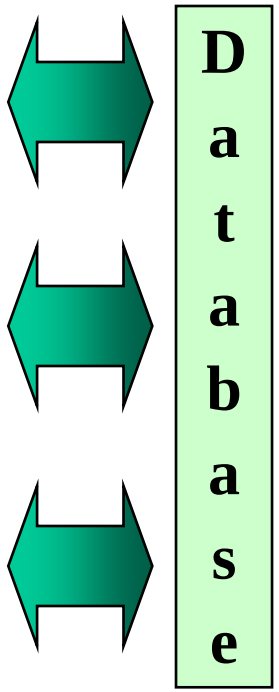
NDA • HRGS

Pellet (U)

Macrostructure analyses
- measurement of physical size

Microstructure analyses
- profilometry

DA
- titration/K-Edge
- TIMS
- ICP-MS/GDMS



Powder (U, Pu, ...)

DA
- titration/K-Edge
- TIMS
- SIMS
- ICP-MS/GDMS

Microstructure analyses
- SEM
- TEM
- EDX

?

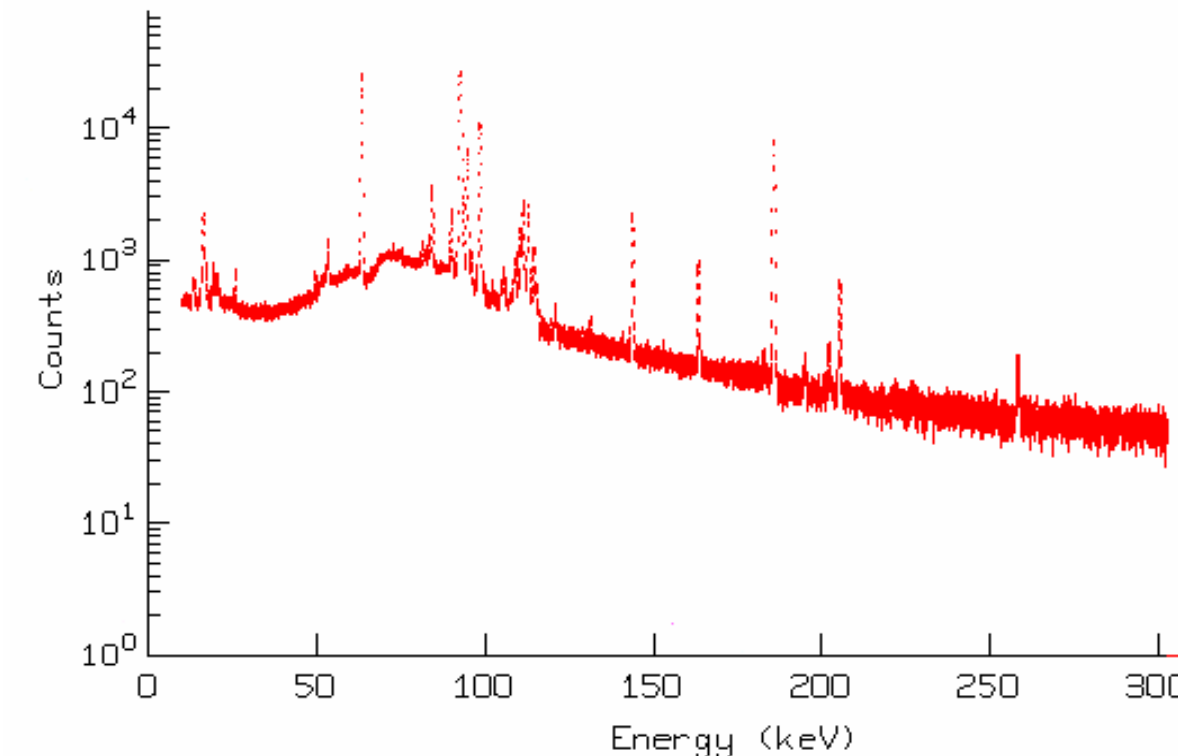
Non-destructive analysis (NDA)

High Resolution Gamma Spectrometry

- plutonium and uranium isotopic composition
- fission products

Neutron Coincidence Counting

- effective ^{240}Pu (spontaneous fission)



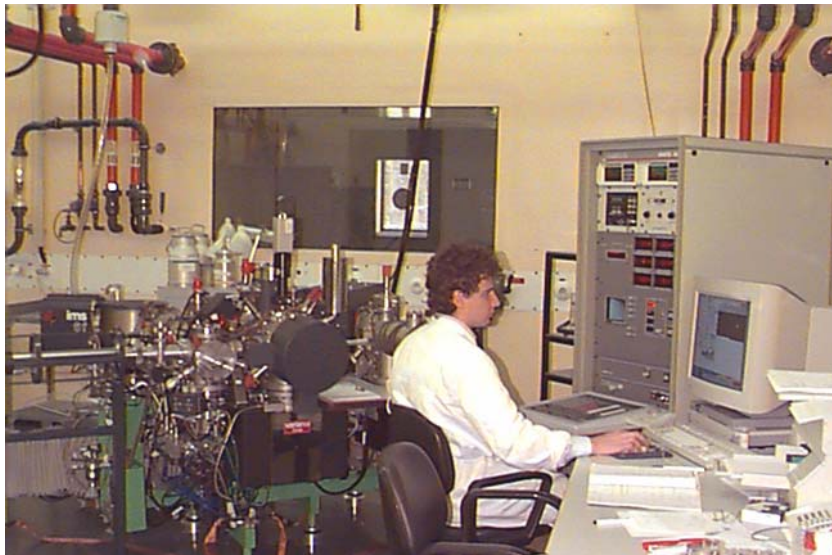
Macro- and microstructure analyses of pellets

- Physical size = dimensions of pellets (height, diameter, central hole size)

- Surface roughness of the pellets using profilometer

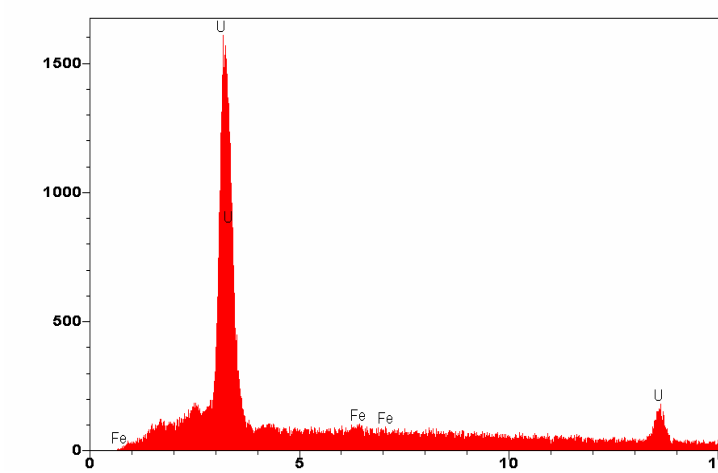
Destructive analysis (DA)

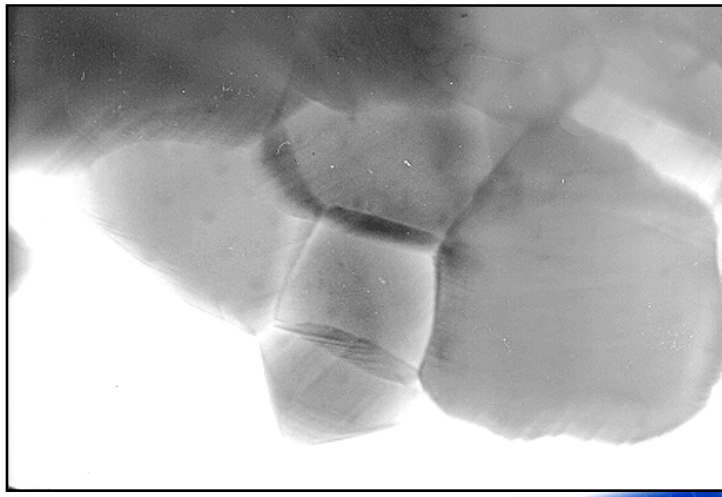
1. Titration and HKED
   - U and Pu content
2. TIMS - (IDMS)
   - isotopic composition (and elemental concentration)
3. ICP-MS and GDMS
   - impurities
4. SIMS
   - isotopic composition and impurities in particles



Microstructure analyses of particles

1. SEM
   - particle size and surface structure
2. EDX
   - chemical composition and elemental distribution
3. TEM
   - internal microstructure (grain size, porosity, dislocations)
   - crystallographic structure





Database

1. Fabrication - fuel manufacturers

- dimensions
- ^{235}U enrichment
- impurities (upper limits, typical range)

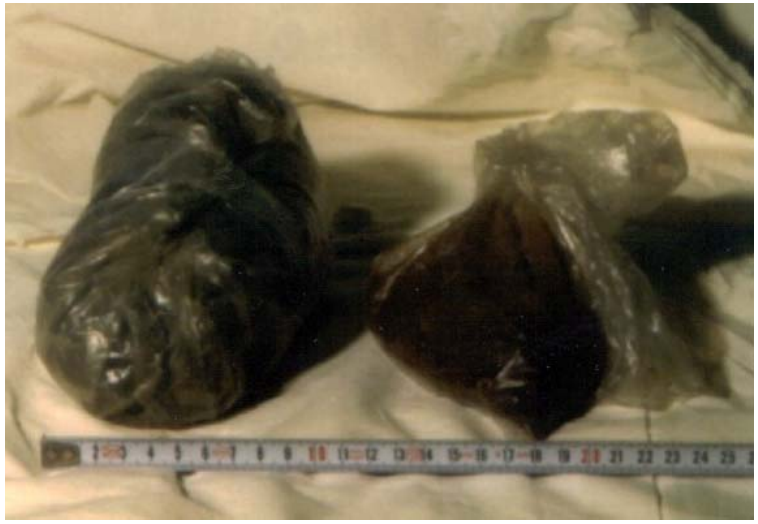
Type	d_o (mm)	d_i (mm)	h (mm)	U_{235}/U_{tot} (w-%)	Er (wt.%)
xxx	11.46 ± 0.02	2.05 ± 0.15	13.5 ± 1.5	2.00 ± 0.05	< 0.001

2. End use - reactors

- commercial reactors - open literature
- research reactors (limited)

3. Fund + experimental data

Fund 22 - Ulm



- small metal can containing 18 g of UO_2-powder
- lead foil containing 1.8 kg of UO_2-powder + 206 U pellets

Powder:

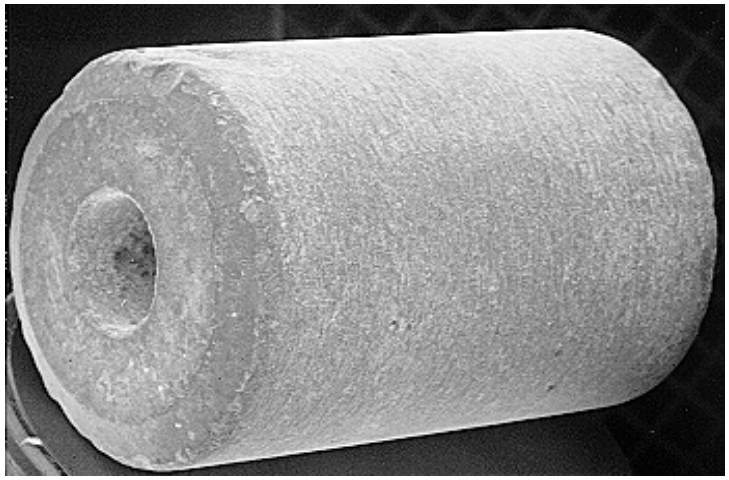
- U-content 69 %

U	234	235	236	238
w-%	0.005	0.711	<0.001	99.285

Element	C	N	Na	S	Cl	K	Fe
ppm	1459	167	254	10680	361	129	353

UO_2-powder was an intermediate product.

Fund 22 - Ulm



Pellets:

U-content 88 % ⟶ UO_2

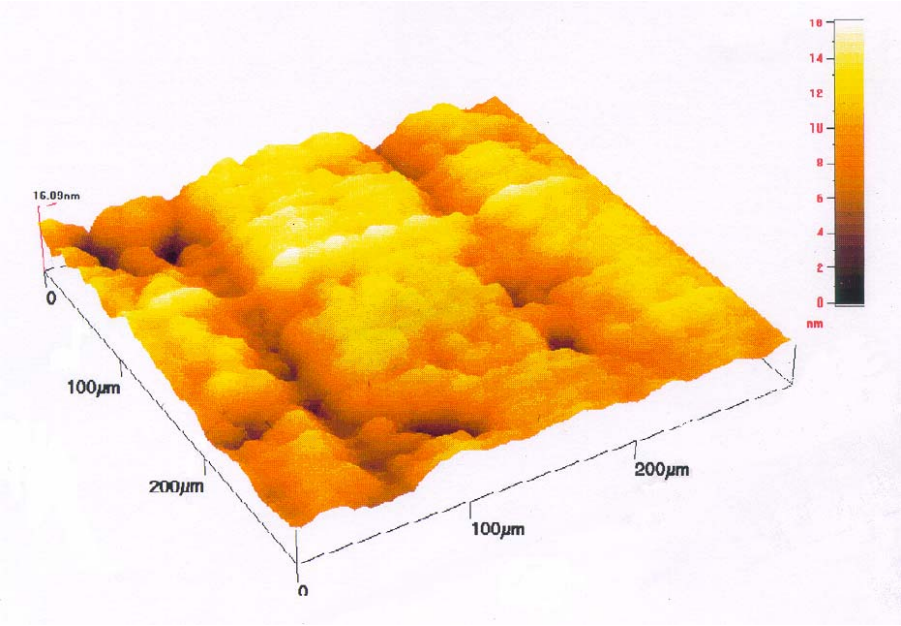
U	234	235	236	238
w-%	0.039	4.38	0.032	95.54

Diameter (mm)	Hole diameter (mm)	Heigh (mm)
7.55	2.43	10.5-12.3

Data Bank VVER-1000

Russia	Ukraine
Balakovo 1-4	Khmelnitski 1
Kalinin 1-2	Rovno 3
Novovoronezh 5	South Ukraine 2-3
	Zaporozhe 1-5

Fund 22 - Ulm

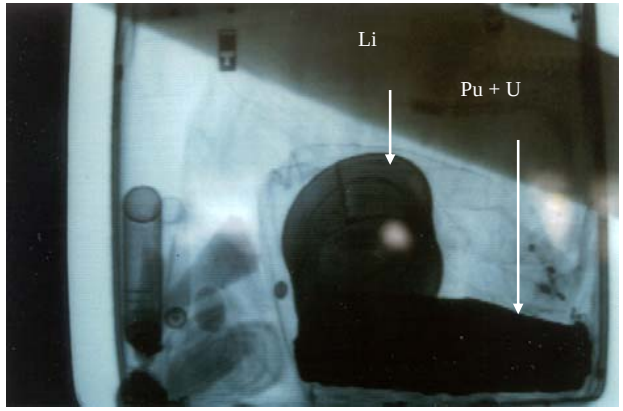


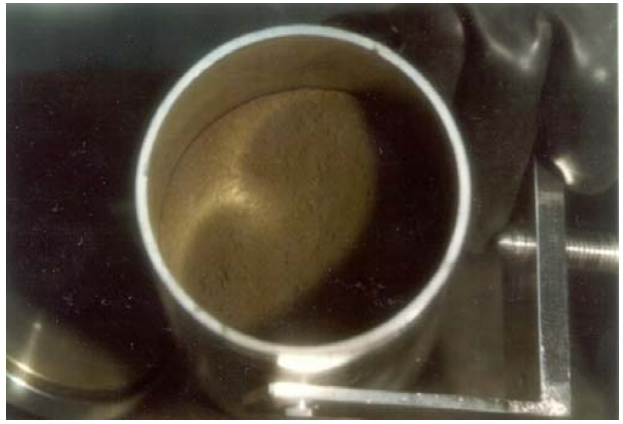
Surface roughness

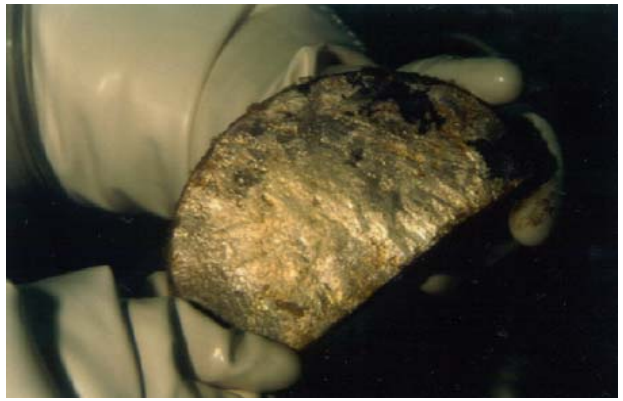
- UMP - wet grinding → smoother surface
- Electrostal-MZ - dry grinding → rougher surface

Measured surface roughness (µm)	
UMP – Kazakhstan	1.7 – 1.9
MZ – Russia	2.3 – 2.5
Pellet – Fund 22	1.87

Pellets were produced in the Ulba Metallurgical Plant, UMP, Kazakhstan.







Fund 19 - Munich

Mixed plutonium and uranium oxides:

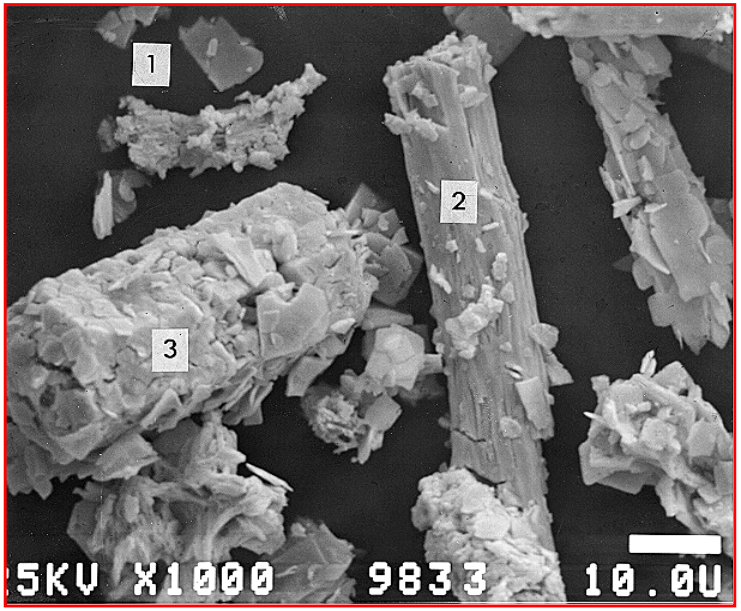
363 g Pu - 121 g U

Pu	238	239	240	241	242
w-%	0.17	87.58	10.78	0.81	0.66

U	234	235	236	238
w-%	0.020	1.60	0.048	98.35

201 g of Li-metal enriched to 89.4 % in ^{6}Li.

Fund 19 - Munich



Powder consists of three particle types:

1) platelets of PuO_2 (80 %)

2) rod-shaped PuO_2 (5 %)

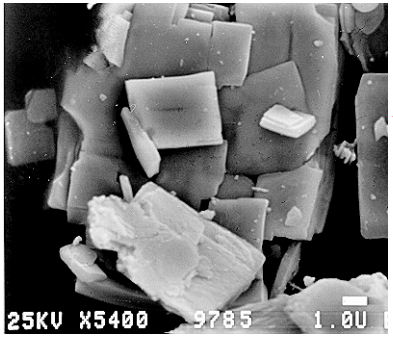
3) hexagonal U_3O_8 (15 %)

	Bulk	Particle 1	Particle 2
$^{240}Pu/^{239}Pu$	0.1226	0.1159	0.1245
Age from the $^{240}Pu/^{236}U^{*}$	(20.6)**	21.5	19.9

* age in years in Feb. 2000

** obtained from the ^{241}Am build-up

Fund 19 - Munich



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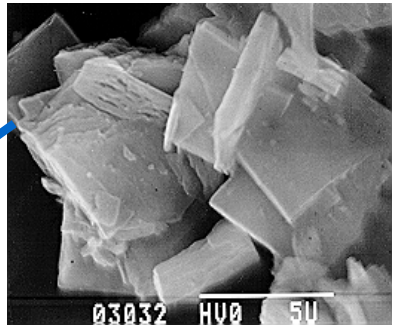
SEM



Reference SM3-1

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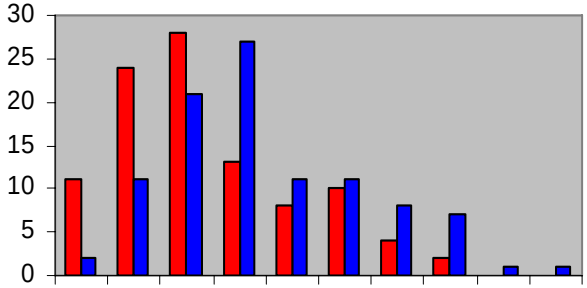
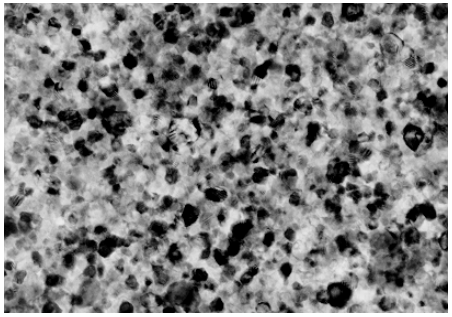
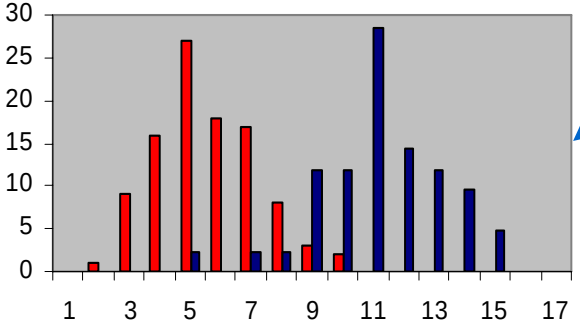
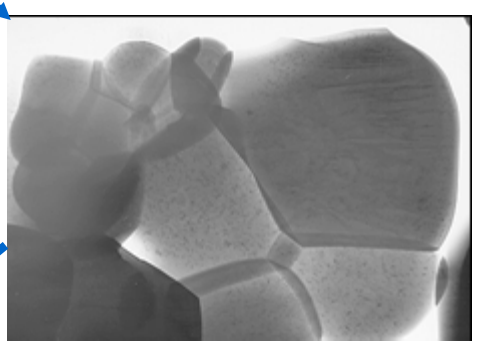




TEM

%





Fund 19 - Munich

1. Plutonium

- weapons-grade
- neither from commercial reprocessing nor from military production
- residues from experiments to develop a MOX process

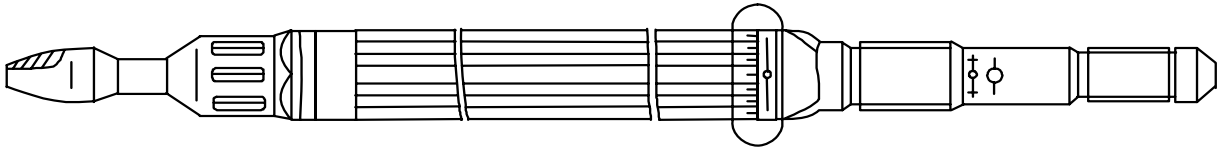
2. Lithium

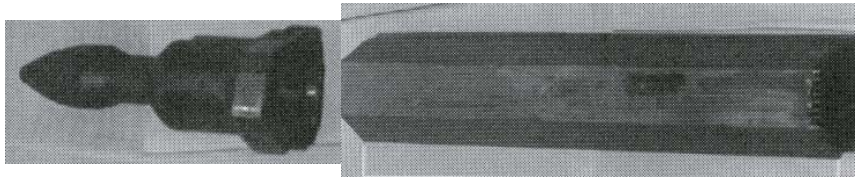
- LiD is used as an explosive in a fusion bomb

Offered material suggests the possible construction of a nuclear weapon.

Origin most probably Russia.

Fund 23 - Rheinhafen



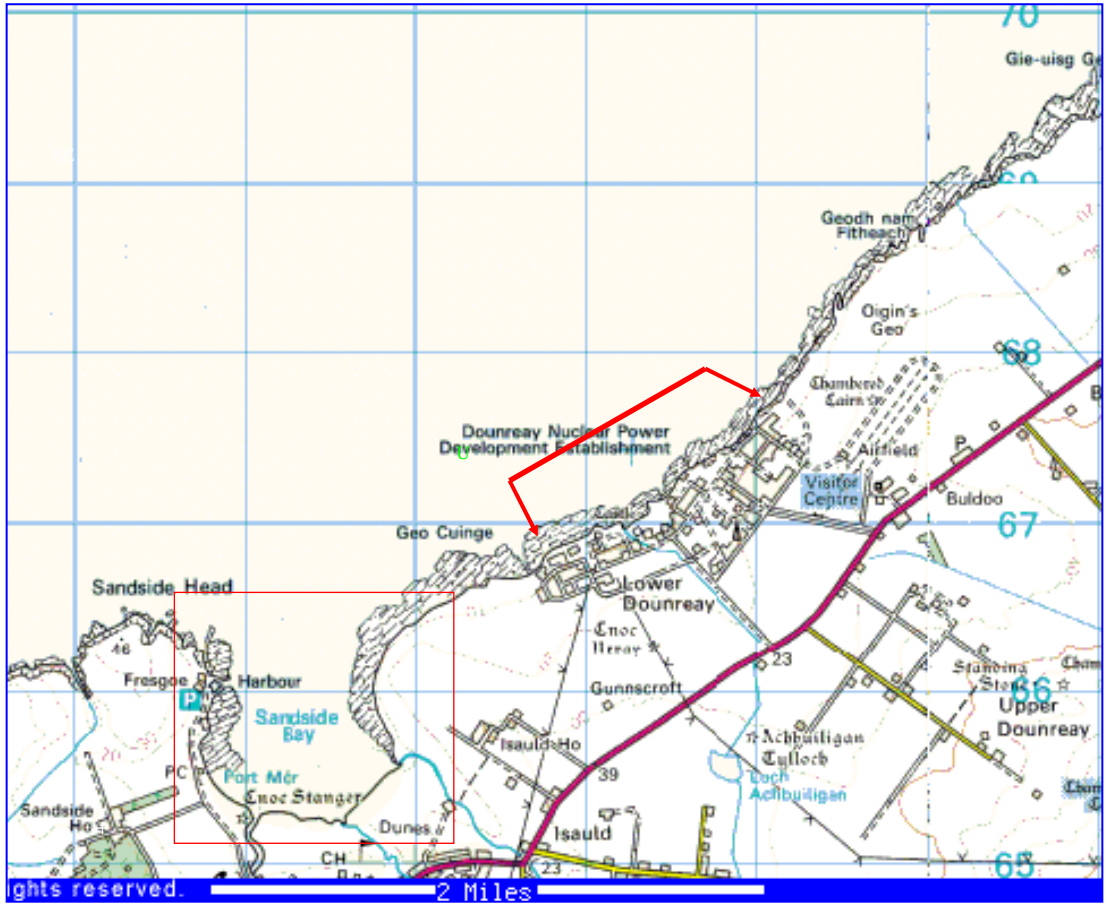


- two pieces of stainless steel
- UO_2 particles (2-10 μm)
- ^{134}Cs and ^{137}Cs

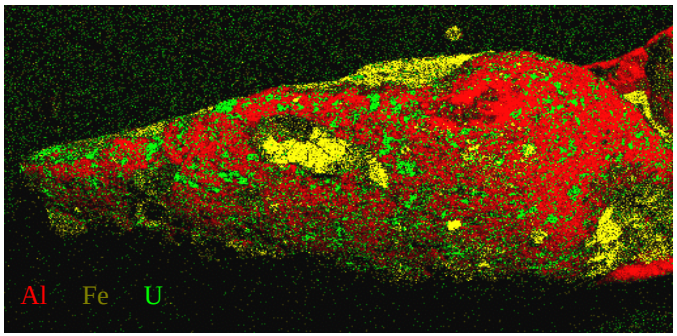
Particle No.	U-235 w-%
1	89.13
2	88.69
3	88.67
4	88.66
5	83.12
6	76.17
7	64.65
8	32.47
9	32.40
10	6.17
11	1.90

- BN-600 fuel assembly
- for testing purposes the fuel is enriched up to 90 % ^{235}U
- e.g. BR-10 in Obninsk

Radioactive particles - Dounreay



- 300 radioactive particles on the foreshore and seabed at Dounreay Establisment, Scotland
- 15 radioactive particles at the beach of Sandside Bay



Radioactive particles - Dounreay

Isotope	SIMS (%)	ICP-MS (%)
^{234}U	1.49 ± 0.02	1.49 ± 0.01
^{235}U	82.9 ± 1.0	82.7 ± 0.9
^{236}U	7.63 ± 0.19	7.56 ± 0.08
^{238}U	7.96 ± 0.21	8.20 ± 0.08

Isotope	SIMS (%)	ICP-MS (%)
$^{239}Pu/^{240}Pu$	6.0 ± 0.3	5.9 ± 0.3
$^{239}Pu/^{238}U$	0.0151 ± 0.0004	0.0143 ± 0.0007

– highly enriched uranium, initial ^{235}U > 90 %

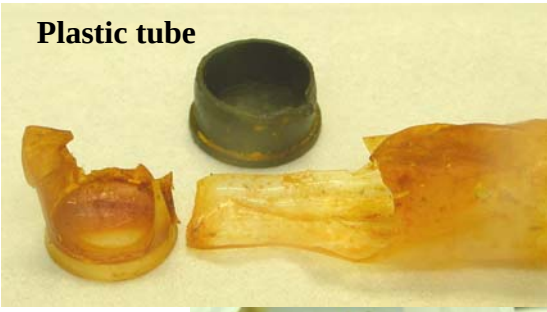
– burn-up 25 - 30 %

– aluminium matrix, Al/U ratio ~ 7

– discharged some 40 years ago

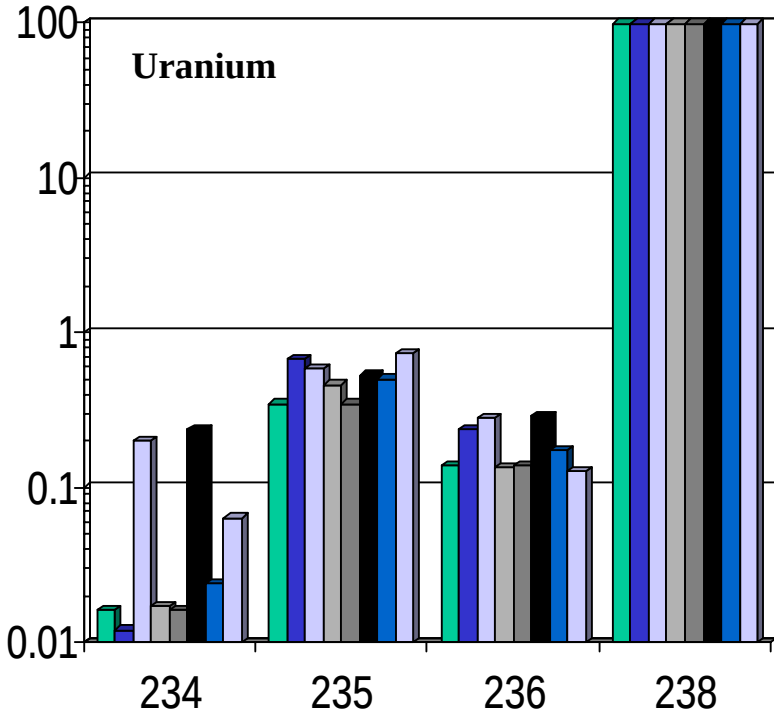
Dounreay Materials Testing Reactor

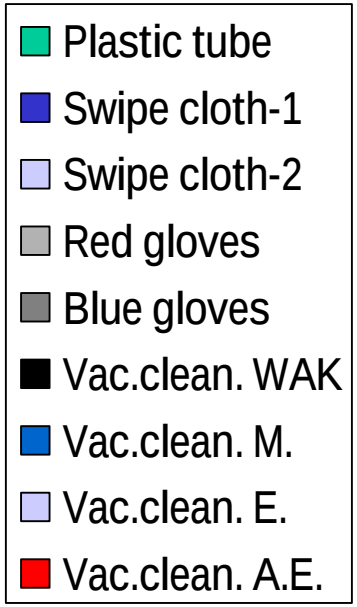
Theft of radioactive waste - Karlsruhe

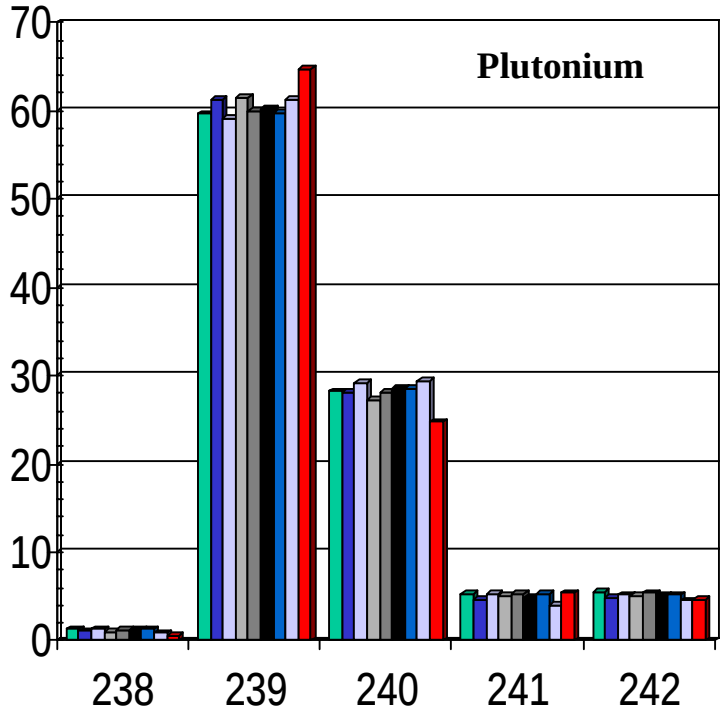


- contamination of an employee was detected in a routine control
- main elements Pu, U, Am and Cs

Theft of radioactive waste - Karlsruhe



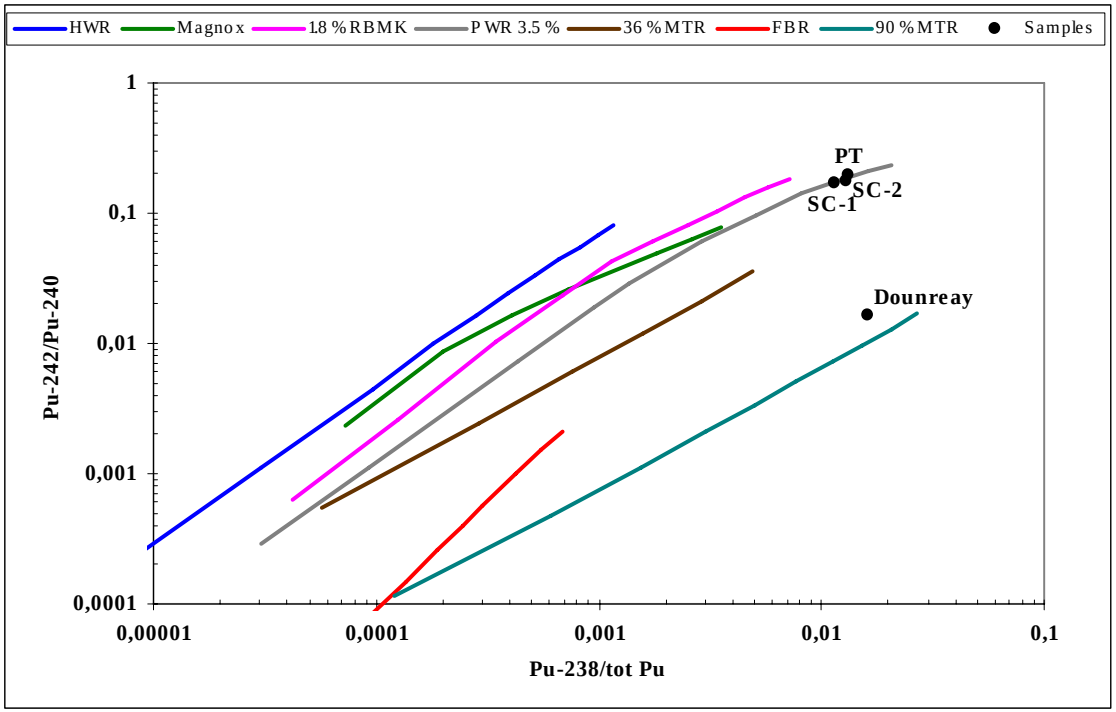




Theft of radioactive waste - Karlsruhe

– The age of the Pu was 12-14 years, i.e. the time of the last reprocessing campaign

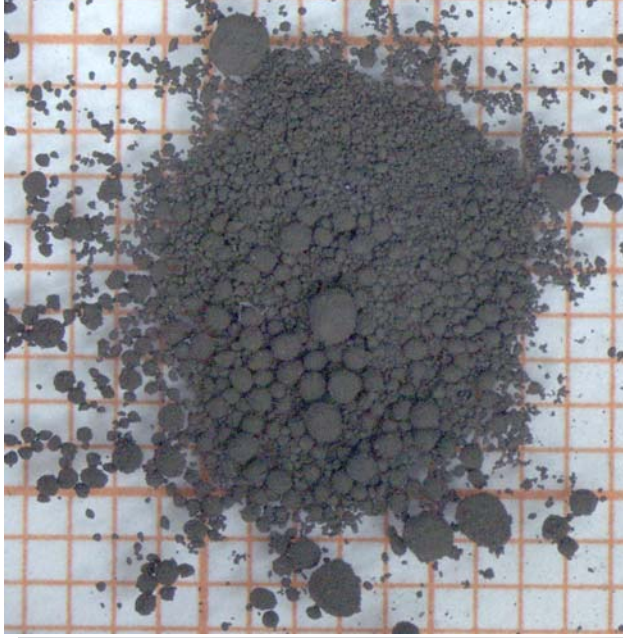
– Pu composition matches with the reprocessed LWR fuel



Joint analysis

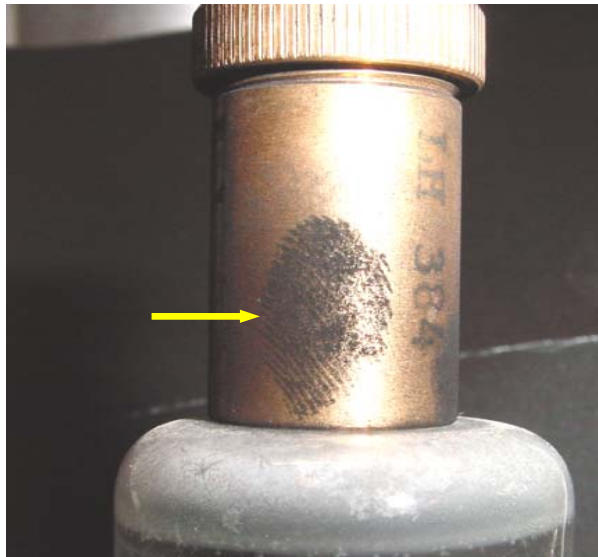
Scope: To give assistance to countries with insufficient analytical capabilities.

1. Czech Republic (2003)
   - Highly enriched uranium
2. Lithuania (2003)
   - low enriched uranium pellets
3. Poland (2005)
   - low enriched uranium pellets and depleted uranium powder



Research & Development

- Age determination (nat.U - HEU and Pu)
- Reactor type determination (Pu)
- Surface roughness (U pellets)
- Oxygen 18/16 ratio for geolocation (nat.U-oxides)
- Impurity patterns for geolocation and production process determination (different U products)
- Fingerprinting contaminated containers



Collaboration

- International Atomic Energy Agency
- Euratom
- International Technical Working Group on nuclear smuggling
- Europol
- Interpol
- Metropolitan Police, Special Branch
- Bundeskriminalamt , Wiesbaden
- Zollkriminalamt, Köln
- Lawrence Livermore National Laboratory